204 Sir W. Ramsay and Mr. F. Soddy. Radioactivity [July 28,

forms nitrous fumes when acted upon by the radium rays or by light.

Becquerel\* mentions the case of mercuric chloride which in presence of oxalic acid is decomposed by light rays, and by radium rays.

- "Experiments in Radioactivity, and the Production of Helium from Radium." By Sir William Ramsay, K.C.B., F.R.S., and Frederick Soddy, M.A. Received July 28, 1903.
  - 1. Experiments on the Radioactivity of the Inert Gases of the Atmosphere.

Of recent years many investigations have been made by Elster and Geitel, Wilson, Strutt, Rutherford, Cooke, Allen, and others on the spontaneous ionisation of the gases of the atmosphere and on the excited radioactivity obtainable from it. It became of interest to ascertain whether the inert monatomic gases of the atmosphere bear any share in these phenomena. For this purpose a small electroscope contained in a glass tube of about 20 c.c. capacity, covered in the interior with tin-foil, was employed. After charging, the apparatus if exhausted retained its charge for thirty-six hours without diminution. Admission of air caused a slow discharge. In similar experiments with helium, neon, argon, krypton, and xenon, the last mixed with oxygen, the rate of discharge was proportional to the density and pressure of the gas. This shows that the gases have no special radioactivity of their own, and accords with the explanation already advanced by these investigators that the discharging power of the air is caused by extraneous radioactivity.

Experiments were also made with the dregs left after liquefied air had nearly entirely evaporated, and again with the same result; no increase in discharging power is produced by concentration of a possible radioactive constituent of the atmosphere.

## 2. Experiments on the Nature of the Radioactive Emanation from Radium.

The word emanation originally used by Boyle ("substantial emanations from the celestial bodies") was resuscitated by Rutherford to designate definite substances of a gaseous nature continuously produced from other substances. The term was also used by Russell ("emanation from hydrogen peroxide") in much the same sense. If the adjective "radioactive" be added, the phenomenon of Rutherford is distinguished from the phenomena observed by Russell.

\* Ibid., vol. 133, 1901, p. 709.

In this section we are dealing with the emanation, or radioactive gas obtained from radium. Rutherford and Soddy investigated the chemical nature of the thorium emanation\* and of the radium emanation,† and came to the conclusion that these emanations are inert gases which withstand the action of reagents in a manner hitherto unobserved except with the members of the argon family. This conclusion was arrived at because the emanations from thorium and radium could be passed without alteration over platinum and palladium black, chromate of lead, zinc dust, and magnesium powder, all at a red-heat.

We have since found that the radium emanation withstands prolonged sparking with oxygen over alkali, and also, during several hours, the action of a heated mixture of magnesium powder and lime. The discharging power was maintained unaltered after this treatment, and inasmuch as a considerable amount of radium was employed it was possible to use the self-luminosity of the gas as an optical demonstration of its persistence.

In an experiment in which the emanation mixed with oxygen had been sparked for several hours over alkali, a minute fraction of the total mixture was found to discharge an electroscope almost instantly. From the main quantity of the gas the oxygen was withdrawn by ignited phosphorus, and no visible residue was left. When, however, another gas was introduced, so as to come into contact with the top of the tube, and then withdrawn, the emanation was found to be present in it in unaltered amount. It appears, therefore, that phosphorus burning in oxygen and sparking with oxygen have no effect upon the gas so far as can be detected by its radioactive properties.

The experiments with magnesium-lime were more strictly quantitative. The method of testing the gas before and after treatment with the reagent was to take  $\frac{1}{2000}$ th part of the whole mixed with air, and after introducing it into the reservoir of an electroscope to measure the rate of discharge. The magnesium-lime tube glowed brightly when the mixture of emanation and air was admitted, and it was maintained at a red heat for three hours. The gas was then washed out with a little hydrogen, diluted with air and tested as before. It was found that the discharging power of the gas had been quite unaltered by this treatment.

The emanation can be dealt with as a gas; it can be extracted by aid of a Töpler pump; it can be condensed in a U-tube surrounded by liquid air; and when condensed it can be "washed" with another gas which can be pumped off completely, and which then possesses no luminosity and practically no discharging power. The passage of the emanation from place to place through glass tubes can be followed by

<sup>\* &#</sup>x27;Phil. Mag.,' 1902, p. 580.

<sup>†</sup> Ibid., 1903, p. 457.

the eye in a darkened room. On opening a stopcock between a tube containing the emanation and the pump, the slow flow through the capillary tube can be noticed; the rapid passage along the wider tubes: the delay caused by the plug of phosphorus pentoxide, and the sudden diffusion into the reservoir of the pump. When compressed, the luminosity increased, and when the small bubble was expelled through the capillary it was exceedingly luminous. The peculiarities of the excited activity left behind on the glass by the emanation could also be well observed. When the emanation had been left a short time in contact with the glass, the excited activity lasts only for a short time; but after the emanation has been stored a long time the excited activity decays more slowly.

The emanation causes chemical change in a similar manner to the The emanation pumped off from salts of radium themselves. 50 milligrams of radium bromide after dissolving in water, when stored with oxygen in a small glass tube over mercury turns the glass distinctly violet in a single night; if moist the mercury becomes covered with a film of the red oxide, but if dry it appears to remain A mixture of the emanation with oxygen produces carbon dioxide when passed through a lubricated stopcock.

## 3. Occurrence of Helium in the Gases Evolved from Radium Bromide.

The gas evolved from 20 milligrams of pure radium bromide (which we are informed had been prepared three months) by its solution in water and which consisted mainly of hydrogen and oxygen\* was tested for helium, the hydrogen and oxygen being removed by contact with a red-hot spiral of copper wire, partially oxidised, and the resulting water vapour by a tube of phosphorus pentoxide. The gas issued into a small vacuum-tube which showed the spectrum of carbon dioxide. The vacuum tube was in train with a small U-tube, and the latter was then cooled with liquid air. This much reduced the brilliancy of the CO<sub>2</sub> spectrum, and the D<sub>3</sub> line of helium appeared. The coincidence was confirmed by throwing the spectrum of helium into the spectroscope through the comparison prism, and shown to be at least within 0.5 of an Angström unit.

The experiment was carefully repeated in apparatus constructed of previously unused glass with 30 milligrams of radium bromide, probably four or five months old, kindly lent us by Professor Rutherford. The gases evolved were passed through a cooled U-tube on their way to the vacuum-tube, which completely prevented the passage of carbon dioxide and the emanation. The spectrum of helium was obtained and practically all the lines were seen, including those at 6677, 5876, 5016, 4932, 4713, and 4472. There were also present three lines of approximate wave-lengths 6180, 5695, 5455, that have not yet been identified.

On two subsequent occasions the gases evolved from both solutions of radium bromide were mixed, after four days' accumulation which amounted to about 2.5 c.c. in each case, and were examined in a similar way. The D<sub>3</sub> line of helium could not be detected. It may be well to state the composition found for the gases continuously generated by a solution of radium, for it seemed likely that the large excess of hydrogen over the composition required to form water, shown in the analysis given by Bodländer\* might be due to the greater solubility of the oxygen. In our analyses the gases were extracted with the pump, and the first gave 28.6, the second 29.2 per cent. of oxygen. The slight excess of hydrogen is doubtless due to the action of the oxygen on the grease of the stop-cocks, which has been already mentioned. The rate of production of these gases is about 0.5 c.c. per day for 50 milligrams of radium bromide, which is over twice as great as that found by Bodländer.

## 4. Production of Helium by the Radium Emanation.

The maximum amount of the emanation obtained from 50 milligrams of radium bromide was conveyed by means of oxygen into a U-tube cooled in liquid air, and the latter was then extracted by the pump. It was then washed out with a little fresh oxygen which was again pumped off. The vacuum tube sealed on to the U-tube, after removing the liquid air showed no trace of helium. The spectrum was apparently a new one, probably that of the emanation, but this has not yet been completely examined, and we hope to publish further details shortly. After standing from the 17th to the 21st inst. the helium spectrum appeared, and the characteristic lines were observed identical in position with those of a helium tube thrown into the field of vision at the same time. On the 22nd the yellow, the green, the two blues and the violet were seen, and in addition the three new lines also present in the helium obtained from radium. A confirmatory experiment gave identical results.

We wish to express our indebtedness to the Research Fund of the Chemical Society for a part of the radium used in this investigation.

<sup>\* &#</sup>x27;Ber.' (loc. cit.).